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ANNUAL REVIEW OF LITERATURE ON RADIOACTIVITY STUDIES OF AIR, WATER AND WASTES, 1955

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ANNUAL REVIEW OF LITERATURE

ON

RADIOACTIVITY STUDIES

OF

AIR, WATER AND WASTES

Edited By

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AND

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Prepared for the Research Committee

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SANITARY ENGINEERING CENTER

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RADIOACTIVITY

Waste Treatment and Disposal

The increased industrial interest in nuclear power was evidenced by a large increase in the number of papers on the problems of radioactive waste disposal. Wolman and Gorman (146, 147, 148) reviewed the sources, characteristics and methods of disposal of radioactive wastes. The dependence of the growth of the atomic energy industry on satisfactory waste disposal methods was recognized. Various estimates of the magnitude and nature of the problem were made (12, 84, 120, 137), and much interest was shown in the sanitary engineering aspects of the atomic energy industry (29, 49, 96).

It was widely assumed that a substantial portion of American power needs will be met by nuclear energy plants [e.g., Jackson (65)]. Terrill, Moeller, and Ingraham (137) predicted that 90 million gallons of highly radioactive fission product wastes would be produced daily in processing nuclear fuel from power reactors built during 1964 alone. Tables and curves were presented from which individual fission-product yields for reactors of any power level and operating conditions could be predicted. Straub (128) emphasized the problems of radioactive waste treatment by pointing out that the permissible limits of radioactive material in water are very low, and also that the different radioactive constituents have individual response characteristics to treatment processes.

A relatively small volume of water wastes from the processing of spent nuclear fuel contains the bulk of the radioactivity in strong

acid or salt solutions. In considering the treatment of such waste for ultimate disposal by clay adsorption, Hatch et al (55, 56) found that high concentrations of hydrogen, sodium, ammonium, nitrate, and fluoride ions were detrimental to the uptake of fission products by the clay. After removal of HNO_3 by distillation and separation of other non-radioactive ions by electrolysis in membrane deionization cells, the fission products were adsorbed on montmorillonite clay. The radioactive clay was then fused into glass beads for ultimate disposal. Higgins and Wymer (59) investigated a method of treating highly radioactive aluminum nitrate wastes. The niobium, ruthenium and zirconium were removed by $Fe(OH)_3$ - MnO_2 scavenging with an efficiency of more than 90 per cent. The $Al(NO_3)_3$ was then converted to dibasic aluminum nitrate by destructive distillation and leached with water at 160° C. to remove Sr, Cs, and rare earths, which were separated by cation exchange for recovery. Co-precipitation methods for the removal of radioactivity from acid aluminum nitrate wastes were studied by Krieger, Kahn, and Straub (79). Ru and Cs were precipitated with $Cu_2Fe(CN)_6$, Sr with $BaSO_4$, Zr and Nb with $ZrO(H_2PO_4)_2$ and the rare earths and Y with $Ce_2(C_2O_4)_3$. A removal of 95 per cent of the activity was obtained with a waste volume reduction of 20:1 at a chemical cost of 1.5 cents/liter.

Walters, Weiser, and Marek (138) described a method of reducing the volume of radioactive wastes. Partial decontamination was obtained in a multicompartment permselective membrane cell followed by final decontamination in a similar cell containing a mixed-bed granular

resin exchanger in the deionization compartments. The mixed-bed exchanger was regenerated by electromigration and the regenerant was evaporated for storage. Swope and Anderson (132) found that a sulfonic acid-type cation-exchange resin can remove 75-80 per cent of the beta-gamma activity due to intermediate levels of mixed fission products from tap water. The resin capacity for activity was more than 260,000 gal./cu. ft. even though 6,000 gal./cu. ft. exhausted its capacity for hardness ions. The principal radioactive elements of the effluent were Ru and Cs.

Dobbins, Edwards, and Gulevich (35) studied one- and two-stage trickling filters for treating laundry wastes containing fission products or other isotopes. Some studies were conducted on actual waste; however, a simulated waste was usually used which consisted of 1,250 ppm citric acid, 2,600 ppm HNO_3, 180 ppm Igepal, 1,725 ppm NaOH, 180 ppm NH_4OH, and 90 ppm Na_3PO_4. The 5-day B.O.D. was 500 ppm, the average pH 4.0, and the gross beta activity usually in the range of 800 to 1100 dpm/ml. of 200-day-old fission products, or in the order of 10^7 dpm/ml. of specific isotopes. Under optimum conditions, percentage removals were: Sr, 69; Ru, 79; Zr, 79; Y, 87; Ce, 97; and fission products, 90. Experiments on the use ofoxidation ponds for removing radioactivity were reported by Steel and Gloyna (124). When mixed fission products were added to 1,200- to 3,000-gallon ponds, 20-25 per cent of the activity was removed for each 5 days detention in the pond. Some smaller-scale experiments on individual isotopes were also made.

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McCauley and Eliassen (93) found that radiostrontium ions can be coprecipitated with $CaCO_3$ as mixed crystals in the cold lime-soda process. Maximum reduction of calcium hardness is essential for a maximum strontium removal of 50 per cent. Repeated precipitations of $CaCO_3$ will remove more than 99 per cent of the radiostrontium.

Manowitz, Richards and Morrigan (92) described the Brookhaven National Laboratory vapor compression evaporator. The 300-gallon-per-hour evaporator reduces the volume of radioactive waste to 1 - 2 per cent of the feed volume.

A summary of the radioactive waste treatment plant operation at Harwell, England, was given by Burns (22). The treatment consists of coagulation and fluocculation followed by effluent discharge to the Thames River at Sutton Courtenay. The British (140) dump to sea radioactive solids contained in concrete blocks. One load of 1,500 tons of material consisted of contaminated apparatus, radiocesium and radiostrontium (13). The volume of wastes from the Shippingsport power reactor will also be reduced by evaporation, according to Simpson and Shaw (119). These wastes from primary coolant, decontamination solutions, flush water, and miscellaneous waste liquid may contain up to 140 μc/rl. The evaporator bottoms will be stored in concrete tanks. The evaporator condensate will be filtered and discharged to the Ohio River with an activity less than 10 μμc/l. Fairbourne, Reid and Kramer (42) summarized the waste disposal operations at the Idaho Chemical Processing Plant. High level wastes are evaporated and stored in tanks. The low level wastes and evaporator condensate are discharged to ground through tile fields.

In order to eliminate indefinite storage of high-level (or concentrated) radioactive waste, discharge to ground was considered (139). Preliminary laboratory studies and the theoretical aspects of underground movement of radioactive waste when injected into wells were reported by Kaufmann, Orcutt, and Klein (70).

Contamination and Decontamination

A radiation survey (46) of a radium clinic showed gross surface contamination by alpha-, beta-, and gamma-emitting contaminants throughout the building and adjacent areas. Evidence of possible minor radiation injury to six of the 10 employees was found but uncertainty of exposure did not permit a correlation with medical findings. Hatfield (57) described the consequences of a mechanical failure in the shut-off rod system of Canada's NRX nuclear reactor. A power surge caused damage resulting in widespread fission product contamination. Special decontamination procedures for equipment, structures and water were described.

Blaschwitz and Judson (18) reported that glass fiber filters were effective (99.99%) in removing particulate matter (0.2 to 0.7 micron in diameter) from waste gas at the Hanford plant. Talboys and Spratt (134) evaluated laundering agents and techniques for the effective decontamination of clothing. Agents which would produce wastes amenable to biological treatment, and techniques, such as dry cleaning, which would minimize the volume of aqueous wastes, were investigated.

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Surveys

Judson and Osmond (67) measured the uranium content and total radioactivity of waters from 77 localities, chiefly in the United States. Uranium values ranged from 0.02 to 460 p.p.b. The high values of both U and total radioactivity occurred in waters from carnotite-bearing beds in the Colorado area. Kawabe (69) measured the radon of 116 hot springs in southern Kyushu. The maximum level found in water was 4470 micro-microcuries per liter, and 93 contained less than 730 μμc/l. The gases from hot springs were not remarkably high in radon content (0.0061 μμc/l.) except for those in an active volcanic region where the radon content varied from 43,000 to 106,000 μμc/l. Stehney (126) found 3-8 μμc/l. each of radium-226 and radium-224 in water of Illinois wells serving a population of over 100,000. This is believed to be the highest authenticated radium content found to date in any drinking water in the U. S. Previous reports of high radium content in drinking water from Frankfurt on Main (Germany) and Pittsburgh, Pa., were not confirmed by Stehney.

A survey of radiation levels at 16 automatically operated radiation monitoring stations in the vicinity of Brookhaven National Laboratory was conducted by Weiss (142). Cooley (30) measured both the numbers and radioactivity of crayfish and other crustacea in the Columbia River. There was no significant change in the biological abundance downstream from the Hanford plant, but a marked decrease in radioactivity of organisms downstream for a distance of 60-100 miles. Young organisms contained more radioactivity than adults and the principal isotope was P-32. Davis and others (33) reported a radiobiological

survey of the Columbia River. Near the reactors, plankton had the highest activity, while further downstream insect larvae and small fish were most radioactive. Relatively high radioactivity, up to 12 μc/kg of scales and 0.53 μc/kg of flesh, was found in whitefish which had migrated from the reactor area and were caught upstream. Krumholtz (80) summarized a comprehensive ecological survey of White Oak Creek, near Oak Ridge, Tennessee. Some radioactivity was found in most of the biota, the most important radioisotopes being P-32, Sr-90, and Cs-137.

Tamevsky and Vassy (135) concluded, from an atmospheric radioactivity survey, that radon accumulates during periods of calm air and is dissipated by winds. They state that rainwater radioactivity is occasionally very much higher than usual, the activity being of artificial origin.

Radioactivity from nuclear weapons tests was reported in rain in India (27), Japan (149), France (1, 47), the United Kingdom (127), and the United States (38, 39, 89, 111, 122). Particulate atmospheric radioactivity of this type was reported in the United Kingdom (127), France (47), and New Zealand (45, 114). Other reports of radioactivity from nuclear weapons came from Japan, including radioactive contamination of the fishing ship "Lucky Dragon" (118) and of its catch (74). Miyake, Sugiura, and Kaneda (94) measured the horizontal and vertical distribution of radioactivity in the sea near Bikini Atoll in June 1954.

In the comprehensive survey of the AEC monitoring program, Eisenbud and Harley (38, 39) estimated cumulated radioactive fallout as of January 1, 1955, ranging from 21 to 120 or an average of 61 mc./sq. mi., exclusive of Nevada. The average Sr-90 fallout was

estimated as 1 mc./sq. mi. The fallout in the United States from the 1955 Nevada tests (89) ranged from 3 to 850 mc./sq. mi. when extrapolated to January 1, 1956. Stewart, Crooks and Fisher (127), basing their conclusions on tests of radioactivity in rain samples and in air samples taken by aircraft, stated that the total external dose in the United Kingdom from all nuclear weapons tests up to May 1955 would reach 1.6 mr. About 70 per cent of the dose is associated with material not yet deposited on the ground. The dose due to dust suspended near ground level is negligible. They estimate an eventual dose, if the present rate of testing continues, of 26 mr. per individual. Peak air concentrations of beta activity (24-hr. averages) are usually about 2.3 μμc per cubic meter, but values as high as 10 μμc/m³ have been observed. Radioactivity originating in Nevada or in Russia was found to be soluble to the extent of 20-30 per cent, while that originating in Eniwetok was soluble to the extent of 55 per cent. Soule (122) reported a marked increase in the radioactivity of surface and drinking water in Massachusetts in April 1953, and Rosenthal (110) discussed the removal of this radioactivity by water treatment plants. Coagulation and sand filtration removed 41 to 52 per cent of the radioactivity.

Effects of Nuclear Weapons

Discussions of the effects of nuclear weapons, especially radiological contamination, were made by the AEC (2), Andrews (9), Cockcroft (28), Lapp (81) and Rosenfeld et al (109). List (86) correlated the meteorological trajectories of bomb debris from the

1952 Nevada tests with fallout patterns in the United States. The maximum observed activity at distances over 2,000 miles was 0.83 μc./square meter due to rainout. The two highest air particulate concentrations observed were 0.6 and 0.003 μc./cu. meter.

Protection against radiological contamination from nuclear weapons was described by Dunning (36), Hill (60), and the Federal Civil Defense Administration (44). The U. S. Atomic Energy Commission (4) described precautions taken to minimize fallout damage during nuclear weapons tests.

The nature of fallout radioactivity was discussed by Maher (90). Iodine-131 and strontium-90 are the isotopes of major hazard. Sax, Gabey, Revinson, and Keisch (116) reported radiochemical analyses of Marshall Islands soil heavily contaminated after the March 1, 1954, test. Chemical procedures for Sr, Ru, Zr, and rare earths are given. There were four papers (71, 72, 73, 129) on the nature and size of the radioactive fallout on the Japanese fishing ship, "Lucky Dragon." Amphlett (8) discussed the fate of radioactive contamination according to the nature of the contaminated ground. Possible treatments of contaminated land are included.

Radiochemical Methods

Rosholt (113) used the polonium daughters for the determination of sources of natural radioactivity. The alpha activity of the polonium, which was precipitated with bismuth sulfide, was measured with a scintillation counter. A combination of decay and aluminum absorption techniques

permitted separate estimation of the radon-222, thorium-232, and lead-210 contents.

Jucker and Treadwell (66), studying the co-precipitation of radium with barium sulfate, found that radium could be quantitatively co-precipitated, and would adsorb on fresh pre-formed barium sulfate. They also found that radium could be co-precipitated with strontium sulfate, but very poorly with calcium sulfate. Cadudal (23) determined atmospheric radon by adsorption on active charcoal, followed by direct beta counting of the charcoal. Sensitivity was 1 μμc/liter when volumes of 100-200 liters were used. Stehney and his co-workers (125) determined breath radon in a similar manner, except that the radon, after adsorption, was desorbed into a proportional counter or ion chamber. For the determination of uranium in urine, Campbell et al (24) wet-ashed and extracted with di-n-butyl orthophosphoric acid. The extract was evaporated, fused, and counted. Accuracy was reported as 84 ± 14% at a level of 0.5 to 5 μμc of U-235.

Lindner (85) compiled a number of radiochemical procedures, and Kahn (68) reported procedures for eluting various radionuclides from soil and determining them. Carter (25) gave methods in use at Harwell for separating old fission products. Strontium-90 is separated as strontium nitrate from 80 per cent nitric acid; cesium-137 by double precipitation as cobaltinitrite, perchlorate, silicotungstate, or double bismuth iodide, followed by solution in sodium hydroxide, and adsorption and elution from a cation-exchange resin; cerium is precipitated as iodate from nitric acid solution; and ruthenium is distilled as

tetrachloride from nitric acid solution containing a suitable oxidant. Ishibashi et al (63, 64) reported on the radiochemical analysis of "the Bikini ashes" and on paper chromatography of radioisotopes. The Bikini ashes were found to contain Ca-45 and U-237 in addition to fission products.

Hahn and Straub (52) gave a procedure for the determination of low concentrations of radiostrontium and radiobarium in water. The limits of sensitivity were 40 μμc/l. for strontium and 100 μμc/l. for barium with a precision of about 10 per cent. Farabee (43) determined radiostrontium and radiobarium in urine. The alkaline earths were first concentrated by precipitation and then wet-ashed. After solution and adjustment of the pH to 5.5, ethylenodiaminetetraacetic acid (EDTA) was added to chelate calcium and magnesium, and the solution was passed through a cation-exchange column. Sr and Ba were adsorbed along with approximately half the Mg and a small part of the Ca. The adsorbed Mg and Ca could be eluted with citric acid and EDTA at pH 5.0. Alkali metals were eluted with 0.5 N HCl, after which the Sr and Ba were eluted with 6 N HCl, dried, and counted.

A combination of solvent extraction and precipitation techniques was used for the separation of rare earths from thorium (83). The same techniques in conjunction with oxidation-reduction were used to determine cerium (48). Two ion-exchange separations of rare earths from each other, using lactic acid eluant, were reported (32, 103). Rare earths were also separated by elution with ethanol and HCl from ion exchange columns (82), and by paper chromatography (105), using 8-hydroxyquinoline solutions.

Swain, Kreiter, and Sheppard (131) reduced water to hydrogen gas, which was then transferred to an ion chamber for determination of tritium content. Wing and Johnston (145) converted tritiated water to acetylene and transferred it, with argon, into a Geiger-Muller tube for counting.

Instrumental Methods

Schmidt (117) described the equipment used at the Chicago South District Filtration Plant for measuring radioactivity in precipitation and in raw and filtered water. The equipment includes internal proportional counters for the measurement of low activities, a Geiger-Muller tube in a flowing stream for continuous monitoring at somewhat higher levels, and battery-powered survey meters for emergency use. Weiss (142) described the instrumentation used in a survey of the Brookhaven area.

Harley and Hallden (53) gave a new graphical method for determining the maximum energy of a beta emitter and for determining impurities in beta emitters of known energy. Suttle and Libby (130) gave a method for the absolute assay of beta radioactivity in thick solids based on the observation that, under closely specified geometric conditions, beta absorption is accurately exponential and, therefore, self-absorption can be described by relatively simple expressions. Pate and Yaffe (101,102) discussed the efficiency of counting in a 4-pi counter, commonly used for primary standardization of radioisotopes.

A number of scintillation counters (31, 34, 50, 51, 78, 99, 100, 144) for alpha and gamma measurements were described. Among these were a combined neutron and absolute alpha counter (50), and devices for measuring radon in air (31) and in living animals (51). A cadmium sulfide crystal (non-scintillation) civil defense monitor was developed (77).

Three ionization chambers (41) for monitoring tritium in air and a balanced ionization chamber (104) for monitoring tritium in air in the presence of moderate amounts of gamma activity were described. Boulanger (19) used an ionization chamber to determine radon at concentrations of 0.1 to 100 μμc/l.

A chemical dosimeter for civil defense purposes, useful in the range 50 to 600 rep, was described by Taplin (136) Brooks and Ehrlich (20) discussed the applicability of the photographic film stocks in retail outlets for radiation surveys in the event of nuclear disaster.

Air Pollution

A number of samplers for air particulates were described (62, 115, 123). Heslep and Bellamy (58) discussed, in general terms, the nature and biological effects of nuclear radiation with special emphasis on airborne radioactive materials and the methods of sampling and measuring them. Waterfield (141) calculated the dosage from a cloud of fission products formed by sudden release from a stack.

Holaday (61) surveyed the radon in American mines. Values for the individual American mines were from 70 to 59,000 μμc/l. Median radon

levels were 600 to 8300 μμc/l. of air, compared to about 1000 μμc/l. in European mines where a high occupational incidence of lung cancer exists. The radiation dose from 1000 μμc/l. was estimated to be 4.3 rem/week to the lung and 72 rem/week to the bronchi. Polonium in the urine of miners was found to vary from 2 to 38 μμc/l., whereas the urine of non-miners had no polonium. Harris (54) found the radon concentration in New York State non-uranium mines varied from 10 to 110 μμc/l.

Biological Effects

Whipple et al (143) discussed personnel protection at the University of Rochester. The major hazards were from inhalation of both alpha- and beta-emitters. A calculation of waste discharge to stream water was also made. Albert and Arnett (5) found that the rate of elimination of thoron daughters from the human lung was insufficient to materially reduce radiation hazards, while Camber (26) found no excess damage from inhaled particulate sulfur-35 as barium sulfate. Kikuchi and co-workers (75, 76) described the metabolism of fission products in the case of the Japanese fishing ship, "Lucky Dragon." These studies included the localization of radiostrontium, the influence of radiostrontium on blood and bone marrow, and the effects of EDTA on the metabolism of radiostrontium and radioyttrium.

A number of papers appeared on the uptake of radioactive materials from soil by plants. Two papers by Long and his co-workers (87, 88) reported on fission product uptake, Rediske and Selders (106) studied cesium uptake, and Romney, Rhoads and Larson (108) measured the

uptake of Sr-90, Ru-106, Cs-137, and Ce-144. The effects of environmental factors on the uptake of strontium-90 (98), of calcium, phosphorus and strontium (37), and of iron (107) were reported.

Applications of Isotopes

The U. S. Atomic Energy Commission (3) summarized the distribution and utilization of isotopes over an eight-year period. A bibliography of over 7000 items, divided into 30 categories, is included. Manov (91) stated that 2000 institutions and 1200 industrial firms have used radioisotopes over the past eight years. He surveyed the applications of these itotopes and the part played by the AEC and by State and Federal agencies in controlling hazards connected with their use.

Both the British at Harwell (10) and the Americans at Oak Ridge (11) have set up research programs on the utilization of reactor wastes. Irradiation sources of cesium-137 (40) and of gross waste fission products (7) were considered. Rather extensive studies (6) have been made on the preservation of meat by irradiation.

The movement of water in soil has been followed (16) with a tritium tracer, and tracer techniques have also been used (112) to study fluid velocity in tubes. The destruction and stabilization of pulp wastes by irradiation appeared economically unfeasible (15). Fullen and O'Connor (21) found the radon content of Westchester County (N.Y.) sewage to be higher than that of natural waters and suggested its use to trace sewage.

Neutron radioactivation analysis was applied by Smales and Salmon (121) to the determination of quantities of rubidium and cesium ranging from 10 to 0.01 μg. North Atlantic sea water was found to contain 120 p.p.b. of Rb, and 0.5 p.p.b. of Cs. Morrison and Cosgrove (95) used activation analysis for the quantitative determination of trace impurities in silicon.

Regulation

A number of articles on the regulation and control of radiation appeared. Bergsma and Rechen (17) discussed the development of the control program in New Jersey. Tabershaw and Harris (133) discussed existing codes and suggested requirements of radiation control legislation. An editorial in the American Journal of Public Health (14) called for assumption by state health officials of their responsibilities in radiological protection.

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